UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
REGION 5

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SUBJECT: Preliminary Remedial Goals (PRGs) for Soil Mirex based on Beef and Milk from Cows in Floodplain Areas, Nease Chemical Company Superfund Site, Salem, OH.

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Summary

Soil mirex preliminary remedial goals (PRGs) are calculated for risk-based targets for consumption of beef and milk products from cows pastured in floodplain areas downstream of the former Nease Company facility (Table 1). The PRGs are based on modeled uptake of mirex in cows through direct exposure (soil ingestion while grazing), and indirect exposure (accumulation of mirex in plants grazed by cattle). Biotransfer of dietary mirex to beef or milk is calculated according to RTI (2005). A range of soil mirex PRGs is presented that bounds uncertainty associated with two variables affecting exposure – mirex bioaccumulation in plants and the fraction of soil in cow's diet. An additional scenario is included to illustrate the effect of providing supplementary clean feed to dairy cows.

Table 1. Soil Mirex Preliminary Remedial Goals for Cattle and Dairy Pasture Based on 10^{-5} Risk-based Targets for Consumption of Beef and Milk, Nease Company Superfund Site, Salem, OH.

Cattle Food Source Assumptions	Product	Soil PRG Range mg mirex/kg soil dw
Graze in and/or provided forage from contaminated floodplains (100 % of total)	Beef	0.6 – 2.8
	Milk	0.3 – 1.4
Graze in or provided forage from contaminated floodplains (26 %) with supplementary clean feed (74 % of total)	Milk	0.5 – 1.6

The combined influence of the variability of soil ingestion and plant bioaccumulation results in a 3- to more than 4-fold range in PRGs. Uncertainty over biotransfer of dietary mirex to beef or milk is not included in the range of PRGs presented here. Instead, a single approach, that of RTI (2005), is used. The rationale for selecting a single biotransfer approach is discussed in Methods and Uncertainty.

The calculated milk-based PRGs are lower than beef-based PRGs because of greater modeled food consumption by lactating cows which results in higher modeled exposure to mirex. Provision of supplementary clean feed to dairy cows, accounting for nearly 75 % of the total diet, has a relatively minor influence on soil PRGs because soil ingestion accounts for much more exposure of cows to soil mirex compared to plant uptake. The same range of soil ingestion values, measured for cattle with supplemental feed, was used in all the scenarios because of data

limitations (soil ingestion of non-supplemented cattle in Central or Eastern United States was not located).

The soil PRGs are based on modeled uptake of mirex in milk and beef during the period of use of potentially contaminated fields. Fields with soil mirex levels at or below the PRGs are expected to limit accumulation of mirex in milk or beef to within or below acceptable risk-based targets, either for actively grazing cattle or for cattle feeding on forage harvested from such fields. The effect of rotation of cattle between contaminated and uncontaminated fields during the grazing period is not included in this memo because this involves a wide suite of options affected by many variables including farm management practices, farm size, herd size, and the relative proportion of potentially contaminated pasture. Fluctuations in milk or beef mirex levels due to seasonal patterns of grazing in potentially contaminated pasture alternating with barn confinement with clean feed are also not included in this memo.

Background

Between 1987 and 1989, mirex was detected in milk and beef from farms adjacent to the Middle Fork of Little Beaver Creek (MFLBC). Alternative drinking water was provided, and fencing installed to prevent access of livestock to contaminated pasture. Mirex has not been detected in either beef or milk since these actions were taken.

The Endangerment Assessment for the Nease Chemical Company (EA 2004) did not evaluate current risks associated with consumption of mirex-contaminated livestock products because fencing successfully eliminated this route of exposure in the farms currently in operation. However, a future exposure scenario was included to evaluate risks associated with breaches of presently fenced areas or establishment of pastures in other floodplain areas not presently farmed. The EA relied on measured mirex levels in beef and milk prior to fencing to estimate potential exposure if livestock are pastured in contaminated portions of the MFLBC floodplain in the future. This approach was used instead of modeled uptake "because of the large degree of uncertainty in bioaccumulation models used to estimate uptake in cows" (EA 2004). This permitted evaluation of potential risks associated with livestock products from cows exposed to similar levels of mirex as the livestock grazing in contaminated areas of the MFLBC before fencing, but does not allow calculation of protective soil mirex concentrations because, without a bioaccumulation model, the relationship between mirex levels in soil and those in beef or milk is uncharacterized.

Purpose

This memo utilizes updated information on modeling biotransfer of organic chemical intake to beef and milk to calculate preliminary remedial goals (PRGs) for soil mirex associated with acceptable risk ranges for human consumption.

Methods

Soil PRGs are calculated by Equation 1:

[1] Soil PRG = $C_{fat\ target}$ / (FIR * BTF_{fat} * (BAF + DF_{soil}))

Where $C_{fat\ target}$ is the target concentration of mirex in beef or milk fat associated with acceptable risk (mg mirex / kg fat), FIR is the daily food ingestion rate of cows on a dry weight basis (kg forage dw / day), BTF_{fat} is the biotransfer factor of mirex to fat ((mg mirex / kg fat) / (mg mirex intake / day)) which simplifies to day / kg fat, BAF is the mirex bioaccumulation factor in aboveground portions of plants on a dry weight basis ((mg mirex / kg plant dw) / (mg mirex / kg soil dw)) which simplifies to a unitless ratio, and DF_{soil} is the dietary fraction of soil on a dry weight basis ingested by grazing cows ((kg soil dw / day) / (kg forage dw / day)) which also simplifies to a unitless ratio. The derivation of Equation 1 is shown in Appendix A.1.

Each element of the equation is discussed below.

Four assumptions are made for calculating soil PRGs: 1) cows graze only in potentially contaminated fields or are fed only with forage harvested from such fields, that is, cows are not rotated between contaminated and uncontaminated pastures; 2) the cow's drinking water is an insignificant source of mirex relative to dietary exposure; 3) inhalation is an insignificant source of mirex to cows relative to dietary exposure; and 4) if provided, supplemental forage is harvested from potentially contaminated fields (on-site source). The method for an additional scenario with provision of supplemental clean feed is described at the end of this section.

The target mirex concentrations in beef or milk fat ($C_{fat\ target}$) are shown in Appendices B.1-B.2, and are calculated from the future reasonable maximum exposure (RME) scenario in the Endangerment Assessment for the Nease Chemical Company (EA 2004) (for details see Beef Target and Milk Target footnotes in Appendices B.1 and B.2).

Default values are used for the food ingestion rate (FIR) – 8 kg /d for non-lactating cattle and 16 kg/d for lactating cows on a dry weight basis. These values have been used in several models (RTI 2005; Birak, et al. 2001; Dowdy, et al. 1996; Travis and Arms 1988), and fall within the ranges reported by the National Research Council for cattle: 7 – 10 kg/d non-lactating and 10 – 25 kg/d lactating (NRC 1987 cited by RTI 2005).

The biotransfer factor of mirex to beef or milk fat (BTF_{fat}) is calculated according to the polynomial regression developed by RTI (2005):

[2] $\log BTF_{fat} = -0.099 \log K_{ow}^2 + 1.07 \log K_{ow} - 3.56$

where K_{ow} is the octanol-water partition coefficient (mirex log K_{ow} = 6.89) (EA 2004; RTI 2005). This results in a mirex BTF_{fat} = 0.13 (mg mirex / kg fat) / (mg mirex intake / day). The RTI (2005) polynomial regression is an improvement over the previous linear regression by Travis and Arms (1988) because it more accurately represents the non-linear relationship between K_{ow} and BTF (BTF decreases at both high and low values of K_{ow} compared to middle values), includes chemicals with a wider range of K_{ow} values in the regression, excludes highly metabolized chemicals that break down after being absorbed by livestock,[1] and calculates steady-state accumulation in livestock for studies of insufficient duration to achieve steady-state concentrations. Birak, et al. (2001) updated the linear regression approach of Travis and Arms (1988) by expanding the underlying data base, evaluating different regression techniques, and reporting 95 % confidence intervals; but retained highly metabolized chemicals, did not adjust short-term studies for steady state accumulation, and did not model the non-linear relationship between K_{ow} and BTF. For these reasons, the Travis and Arms (1988) and Birak, et al. (2001) approaches are not used in this memo for calculating PRGs.

Another approach by Dowdy, et al. (1996) for calculating BTF from a regression based on polar-corrected normal path first-order molecular connectivity index ($^1X_{pc}$), a type of quantitative structure-activity relationship method (QSAR), is not used because the results of the technique have been shown to be sensitive to the assumptions used in calculating $^1X_{pc}$ (Appendix B in RTI 2005). The performance of this approach in predicting BTF of an expanded set of chemicals (RTI 2005) was significantly poorer compared to the performance reported in the original paper (Dowdy, et al. 1996). However, the PRGs obtained with this alternative approach are shown in Appendices B.1-B.3 for comparative purposes.

Mirex BTF could also be calculated from two studies in which cows were fed mirex (Bond, et al. 1975; Dorough and Ivie 1974). These empirical results are not used in this memo because of the uncertainties associated with the analytical methods for mirex at these early dates. Since no mirex BTF studies were located that use updated mirex analytical methods, BTF in this memo relies on a general regression based on BTF studies performed with a large number of chemicals.

A range of plant bioaccumulation factors (BAF) is used to account for uncertainty. One estimate is based on a regression between BAF and K_{ow} reported by Travis and Arms (1988) for aboveground plant parts:

[3] $\log BAF = -0.578 \log K_{ow} + 1.588$

which results in a mirex soil-to-plant BAF of 0.004 (mg mirex / kg plant dw) / (mg mirex / kg soil dw).

Another estimate is based on a regression between BAF and the normal path first-order molecular connectivity index (^1X) reported by Dowdy and McKone (1997):[2]

[4] $\log BAF = -0.204\ ^1X + 0.589$

[1] Specifically, oxidizable and hydrolyzable chemicals were excluded because their BTFs were shown to be outliers compared to more stable chemicals. Chemicals that are metabolized into known persistent products that were reported in bioaccumulation studies were retained, for example, conversion of DDT to DDE, aldrin to dieldrin, or heptachlor to heptachlor epoxide (RTI 2005).

[2] The equation in Dowdy and McKone (1997) uses the polar-corrected normal path first-order molecular connectivity index ($^1\chi_{pc}$), but no polar corrections were made for mirex, so the equation for mirex is properly represented by the normal path first-order molecular connectivity index ($^1\chi$).

where mirex $^{1}X = 9.5$ (Dowdy and McKone 1997), which results in a mirex soil-to-plant BAF of 0.045 (mg mirex / kg plant dw) / (mg mirex / kg soil dw).

One study of mirex accumulation in crop seedlings was performed (de la Cruz and Rajanna 1975), but is not used for the BAF in this memo because of the uncertainties associated with the analytical methods for mirex at this early date. An additional uncertainty is that it was a laboratory study performed with spiked soils, which means that the recently added mirex in the laboratory soil may be more bioavailable compared to "aged" mirex that has resided in soil for decades. Since no mirex BAF studies were located that use updated mirex analytical methods, the BAFs in this memo relies on general regressions based on BAF studies performed with a large number of chemicals.

The two BAF approaches used in this memo result in an order-of-magnitude range in mirex soil-to-plant BAFs, which is consistent with the range of BAFs observed even for well-studied individual organic chemicals (McKone and Maddalena 2007).

The range of values for the fraction of soil in a cow's diet (DF_{soil}), 1.4 to 3.8 % on a dry weight basis, is from a study of dairy cattle performed in Michigan (Fries, et al. 1982) selected because the conditions are expected to be representative for Ohio. The range encompasses two treatments with non-lactating cattle (yearling heifers and dry cows) provided supplemental feed with access either to pasture (mean 1.4 to 2.4 % soil dietary fraction) or to unpaved lots with sparse vegetation (mean 1.6 to 3.8 % soil dietary fraction) (Fries, et al. 1982). Unfortunately, the lactating cows in the study were not allowed access to either pasture or lots with sparse vegetation. The overall range of the mean values for the two treatments is used in this memo because livestock soil dietary fraction is inversely related to forage availability – lowest in spring during vigorous growth, and highest in fall or winter when forage is sparse and cows are forced to graze close to the ground (Fries 1996). Similarly, soil dietary fraction is expected to be higher on degraded pastures than on productive pastures, and higher in dry years. The low end of the overall range (1.4 %) is representative of the expected 1-2 % soil dietary fraction for lactating dairy cows fed a supplemental concentrate under normal management conditions (Fries 1996). The upper end of the overall range (3.8 %) represents soil ingestion associated with either poorly managed pasture, dry years, or no provision of supplemental feed. For example, the average soil dietary fraction of dairy cattle without supplemental feed in New Zealand ranged from 4 to 8 % (Healy 1968 cited in Fries, et al. 1982). The relevance of New Zealand range conditions to Ohio is uncertain, so the Healy (1968) values are not used in this memo. Unfortunately, soil ingestion values were not located for Eastern or Central U.S. pastured cattle without supplemental feed, so the high end of the range for Midwestern cows with supplemental feed but grazing on sparsely vegetated lots serves as a surrogate.

Milk production of dairy cows is usually greater when provided supplemental feed in comparison to pasturing without supplement. An additional scenario is included to evaluate the effect of provision of clean (mirex-free) feed to cows also grazing in contaminated pastures. The soil dietary fraction is not adjusted because the values are based on studies of cattle provided supplemental feed. The plant BAF is adjusted to account for the inclusion of clean feed in the total diet, that is, the adjusted BAF (BAF_{adj}) represents the bioaccumulation of mirex in the combined pasture forage and supplemental clean feed diet of the cattle. The modified equations for calculating BAF_{adj} and soil PRG are shown in Appendix A.2. Mean values of 25.7 % pasture forage, and, correspondingly, 74.3 % supplemental clean feed by dry weight are based on 11 dairy farms in NY and PA (range of 9-35 % pasture) (Soder and Muller 2007). The dairies selected for this study provided partial total mixed rations (pTMR), which refers to combining pasture with supplemental feed consisting of a variable mix of silage (fermented high moisture fodder), straw, protein (soybean products), grains, molasses, minerals, and so forth. A different type of supplement using concentrate was not included in the Soder and Muller (2007) study.

Results

The calculated soil preliminary remedial goals are shown in Table 1 for beef and milk targets based on 10^{-5} risk-based targets. A mirex soil PRG range is presented that bounds uncertainty associated with two key variables – mirex bioaccumulation in plants and the fraction of soil in cow's diet. The combined influence of the two variable factors results in a more than a 4-fold range for either beef- or milk-based PRGs. Milk-based PRGs are about one-half of the beef-based PRGs because lactating cows are modeled with twice the food ingestion rate, and correspondingly greater mirex intake rate, compared to beef cattle. The additional scenario of adjusting the plant mirex BAF to account for a mixed diet of pasture forage and supplemental clean feed had only a minor effect on the soil PRGs because of the predominant influence of soil ingestion on cattle exposure to soil mirex. However, with dilution of mirex in the combined forage and feed, the PRG range decreased to approximately 3-fold.

Soil PRGs corresponding to alternative risk-based milk and beef targets are included in Appendices B.1-B.3 for comparative purposes.

The more than 4-fold range in soil PRGs for any single milk or beef target is the combined result of a nearly 3-fold range in soil dietary fraction and a 10-fold range in plant bioaccumulation. This indicates that mirex exposure of cows is influenced more by incidental soil ingestion than by grazing on vegetation at the same location. Soil ingestion also has been reported to be the major source of PCBs in cow's milk (Mamontova, et al. 2007).

The results of one set of alternate mirex diet-to-beef or milk biotransfer factors (Dowdy, et al. 1996) are shown in Appendices B.1-B.3. The alternative BTF_{fat} are higher than the RTI (2005) value, and therefore result in lower soil PRGs. The alternative approach is considered to be less reliable compared to RTI (2005), but is included for transparency. This is discussed further under Uncertainty.

Uncertainty

There are a number of uncertainties associated with the human health risk assessment that is the basis for the milk and beef mirex targets. For a complete discussion of these uncertainties, including uncertainties related to the RME scenario and risk distribution, the Endangerment Assessment should be consulted, particularly section VIII.D (EA 2004).

Multiple sources of uncertainty affect each of the remaining elements of the equation for calculating soil PRGs: food ingestion rate, plant bioaccumulation, soil dietary fraction, and diet-to-beef or milk biotransfer (Equation 1). As discussed below, the uncertainties associated with food ingestion rate and plant bioaccumulation are not likely to appreciably affect the soil PRGs – food ingestion rate because the range of variation is small, and plant bioaccumulation because it has less influence on total mires exposure compared to soil ingestion. Soil dietary fraction appears to be the primary factor determining cattle exposure to mirex. The diet-to-beef or milk biotransfer factor is based on the best available method, which incorporates several important improvements over previous methods, but it still has relatively high uncertainty for mirex because biotransfer studies have not been performed with updated mirex analytical methods.

The food ingestion rates (FIR) in this memo consist of fixed values for lactating and non-lactating cattle. The FIRs are approximately middle values within the ranges given by the National Research Council (NRC 1987 cited by RTI 2005). Based on the NRC ranges, FIR for non-lactating cattle may range from -12 % to +25 % of the 8 kg/d value in this memo, and the FIR for lactating cows from -38 % to +56 % of 16 kg/d. Since these are small ranges (well under 2-fold) compared to the other variables, and the extremes of the ranges are less likely than values near average, uncertainty over FIR is not a significant source of uncertainty for soil PRGs.

The calculated mirex soil-to-aboveground plant bioaccumulation factor (BAF) is highly uncertain, with a modeled 10-fold range. Even experimental results for well-studied organic chemicals show large variability in BAFs, and differences among models show even greater variability (McKone and Maddalena 2007). BAF varies by chemical, soil properties, plant species, and plant part. Additional uncertainty may be that BAF is not constant for a particular combination of chemical, soil, and plant, but may exhibit an inverse relationship with the initial soil concentration, for example, as reported in a field study of PCBs (Pier, et al. 2002). This effect is evident in the sole laboratory study of mirex bioaccumulation in plants (de la Cruz and Rajanna 1975) in which the mean BAF to all aboveground parts of all species (crop seedlings) on field soils ranged from 0.10 to 0.34 (mg mirex / kg plant dw) / (mg mirex / kg soil dw) in soils with mirex concentrations ranging from 3.5 to 0.3 mg/kg, respectively (calculated from their Table 1). Each of the 4 crops included in the experiment exhibited different BAFs, and, within each crop, mirex concentrations were highest in the lower stems of the seedlings, and decreased significantly in leaves and growing tips. The range of mean BAFs for all crop leaves and growing tips was 0.05 to 0.23 in the field soil (from their Table 1). This means that the choice of which crops, which aboveground plant parts, and which soil concentrations to include greatly affects the mean BAF calculated from this one experiment (the researchers also repeated the entire experiment with another soil, a loamy sand).[3]

Another uncertainty is that the availability of soil mirex for volatilization, an important step in exposure to plant foliage,[4] may decrease over time as the soil mirex "ages" and becomes more

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[3] The BAF from the de la Cruz and Rajanna (1975) study reported by Travis and Arms (1988) in their compilation of BAF values from published literature was apparently calculated by selecting the following sources of data: leaves and growing tips of all species, high soil mirex concentration treatment, and both types of soil. Much different BAFs are obtained if data for stems are included, or data from treatments with lower soil concentrations. Dowdy and McKone (1997) give a similar but not identical BAF in their compilation for the same study, but the detailed basis of their calculation is not apparent.

[4] Despite the relatively low volatility of mirex, volatilization from soil is the main exposure pathway to plant foliage. The mirex log octanol-water partition coefficient (K_{ow}) of 6.89 (RTI 2005) is high enough that translocation of mirex from roots to aboveground plant parts is expected to be negligible (Figure 2 in Collins, et al. 2006). The

tightly bound within the soil matrix. For example, the toxicities of DDT and dieldrin to insects decreased as the pesticides aged in soil, but there were only small decreases in the soil chemical concentrations over the same time period, which indicates that the bioavailability of the soil residues decreased over time (Robertson and Alexander 1998). Mirex was added to the soils used in the de la Cruz and Rajanna (1975) laboratory experiment, and therefore the study results may overstate bioaccumulation in plants from mirex that has resided in soil over long periods of time. A complication is that the degree of sequestration with aging is affected by the soil type, in one experiment greatest in muck soil, intermediate in loam, and lowest in sandy loam (Edwards, et al. 1957 cited in Robertson and Alexander 1998). This indicates possible influences of soil organic matter content and soil texture on sequestration. The potential significance of aging of mirex residues in the MFLBC floodplain therefore is partly dependent on the soil types.

The analytical methods for mirex at the time of the de la Cruz and Rajanna (1975) study are not as accurate as current methods, so there is additional uncertainty over the quality of the plant uptake data of this study.

Although the uncertainty over plant bioaccumulation of mirex is large (10-fold), this does not have a commensurate impact on the soil PRG calculations. The reason is that the predominant source of mirex to grazing cattle is through direct soil ingestion.

The diet-to-beef or milk biotransfer factor (BTF_{fat}) used in this memo is calculated by the polynomial regression by RTI (2005), which includes several important improvements over previous regression methods (see Methods). The RTI (2005) polynomial regression performs well in predicting transfer of chemicals into beef or milk fat, with a reported $r^2 = 0.83$, which means that the regression accounts for 83 % of the variation in biotransfer of different chemicals (excluding highly metabolized chemicals). The RTI (2005) approach was developed in response to external peer review of BTFs based on Travis and Arms (1988) (USEPA 2005a).

A completely different approach is to estimate biotransfer with a mechanistic model that includes the processes in an animal that affect absorption, transformation, growth dilution, and excretion of chemicals (Hendriks, et al. 2007). In an extensive comparison, Hendriks, et al. (2007) reported their model performed much better in predicting biotransfer to beef or milk compared to previous linear regression methods, but, for stable chemicals, their model predicted "equally well" as the polynomial regression by RTI (2005). This provides independent support for the RTI (2005) approach.

The linear regression by Travis and Arms (1988) predicts much higher biotransfer of mirex, and consequently lower soil PRGs compared to the RTI (2005) approach (data not shown).

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mirex log octanol-air partitioning coefficient (K_{oa}) of 8.57 (Paterson, et al. 1991) is associated with foliage uptake dominated by gas phase/vegetation equilibrium partitioning and kinetically limited gaseous deposition (Figure 1 in McLachlan 1999; Figure 3 in Collins, et al. 2006), in other words, volatilization from soil. Even though the vapor pressure of mirex is very low, the air-to-leaf bioconcentration factor is very high: 1.2×10^7 on a volumetric basis in azalea leaves (Bacci, et al. 1990). Plants are also exposed to soil mirex through transfer from dust adhering to leaf surfaces, but this is not a major pathway for plant uptake of mirex because particle-bound deposition does not become the dominant pathway for plant uptake until log K_{oa} exceeds 11 (McLachlan 1999, Collins, et al. 2006). However, dust blown onto vegetation can be an important source of soil contaminants to grazing livestock, even if not absorbed by the plant (Beresford and Howard 1991), and may account for much of the soil ingestion by cattle.

However, Travis and Arms (1988) included both stable and highly metabolized chemicals in their regression. Since the metabolized chemicals generally have low K_{ow}, and low biotransfer (because of high rates of elimination), this skewed the K_{ow} – BTF regression to a steeper slope than appropriate for more stable chemicals. Also, Travis and Arms (1988) had few data for high K_{ow} chemicals compared to later compilations. The net result is that the Travis and Arms (1988) linear regression tends to over-predict biotransfer of high K_{ow} chemicals (RTI 2005), such as mirex. The Travis and Arms (1988) mirex BTF and the associated soil PRGs therefore are not considered reliable.

Birak, et al. (2001) supplemented the empirical database used by Travis and Arms (1988), including increasing the database for high K_{ow} chemicals, and investigated using both ordinary least squares regression and geometric mean regression, but did not delete non-stable chemicals, adjust for steady state, or model a non-linear relationship between K_{ow} and BTF. Not surprisingly, the Birak, et al. (2001) regressions perform less well in comparison with RTI (2005), with $r^2 = 0.46$ to 0.50 for milk and beef BTFs, respectively. A unique feature is Birak, et al. (2001) provided 95 % confidence intervals for their regressions. The range of soil PRGs calculated with their upper and lower 95 % confidence interval regressions spans 4 to 5 orders of magnitude (not shown). Much of this reflects forcing linear regressions to a non-linear relationship, combining data on stable and unstable chemicals, and the differences between geometric mean and ordinary least squares regression approaches, but part of this is also indicative of the large variability observed between K_{ow} and BTF.

The regression based on molecular connectivity index by Dowdy, et al. (1996) results in biotransfer estimates for mirex that are intermediate between those of Travis and Arms (1988) and RTI (2005). Dowdy, et al. (1996) reported very good performance of their approach, with $r^2 = 0.90$ and 0.89 for beef and milk. However, the performance declined when applied to an expanded database, with $r^2 = 0.54$ and 0.55 (Appendix B in RTI 2005). For this reason, the soil PRGs calculated with the Dowdy, et al. (1996) BTF are considered more uncertain than PRGs calculated with the RTI (2005) BTF.

Two empirical studies of mirex uptake in cattle have been published (Dorough and Ivie 1974; Bond, et al. 1975). Both studies measured mirex in cattle fat after depuration, 28 and 10 days after cessation of mirex exposure, respectively. To accurately determine biotransfer, mirex levels in fat should be measured on the last day of exposure. However, for comparative purposes, the BTF_{fat} with 10 days depuration is 0.12 for 16 mg mirex / day intake, and 0.38 for 0.16 mg mirex / day intake (calculated from Bond, et al. 1975). The BTF_{fat} with 28 days depuration is 0.013 for 4 mg mirex / day intake (calculated from Dorough and Ivie 1974), which indicates substantial elimination of mirex over time when exposure is stopped. The first value is consistent with the 0.13 BTF_{fat} based on RTI (2005), and the second approaches the 0.48 BTF_{fat} based on Dowdy, et al. (1996), but both of the empirical values are uncertain because of uncertainties associated with the analytical methods available at that time for mirex, and the time lags between cessation of mirex exposure and collection of fat samples.

For milk, three BTF_{fat} values may be calculated from the two studies – 0.13 for 16 mg mirex / day intake (calculated from Bond, et al. 1975), 0.36 for 4 mg mirex / day intake (calculated from Dorough and Ivie 1974), and 1.56 for 0.16 mg mirex / day intake (Bond, et al. 1975). Similar to plant BAF, BTF exhibits an inverse relationship with exposure. Again, the first value is consistent with the 0.13 BTF_{fat} based on RTI (2005), and the second with the 0.33 BTF_{fat} based on Dowdy, et al. (1996), but all 3 values are uncertain because of uncertainties associated with the analytical methods available at that time for mirex. Another consideration is RTI (2005) excluded the results of the 0.16 mg mirex / day intake treatment of the Bond, et al. (1975) study because the mirex concentrations in milk were close to the detection limit, and therefore considered unreliable (see their Table F-2).

The RTI (2005) polynomial regression for estimating mirex BTF is selected as the sole approach for this memo because it incorporates several advances in data analysis compared to earlier approaches, is based on a larger chemical database than earlier efforts, exhibits high performance (high r^2) in predicting biotransfer for a wide range of chemicals, and is supported in that the predictive performance of the RTI (2005) method for stable chemicals is equivalent to that of an independent mechanistic modeling approach (Hendriks, et al. 2007).

The main uncertainty is that mirex biotransfer studies performed with updated mirex analytical methods are not available to confirm (or contradict) the RTI (2005) mirex biotransfer prediction. The existing mirex feeding studies with cattle, performed with older and more uncertain mirex analytical methods, support the RTI (2005) mirex BTF estimate in some treatments, but indicate higher biotransfer in other treatments.

Cattle soil ingestion is a key variable for calculating mirex PRGs for floodplain pasture because it is the main exposure pathway for mirex uptake. The uncertainty associated with soil ingestion is intertwined with provision of supplemental feed, pasture condition, and lactation status. Supplemental feed and pasture condition are influenced by farm management practices, which, in turn, are affected by broader economic trends and farm policies.

The 1.4 to 3.8 % range of mean cattle soil ingestion values is based on non-lactating cattle in the Midwest provided supplemental feed and grazing in pasture or sparsely vegetated lots, respectively (Fries, et al. 1982). Data for lactating cows would have been preferred because of the greater food intake, but this is probably not an important source of uncertainty because soil ingestion is expressed as the percent of diet, not total soil intake. As discussed under Methods, the low end of the range is consistent with the soil ingestion expected with normal dairy management including supplemental feed, which indicates the data are reasonably representative for lactating cows. The high end of the range approaches the 4 to 8 % range reported for grazing cattle without supplementary feed in New Zealand (Healy 1968 cited in Fries, et al. 1982), which indicates the high end partly captures potential soil ingestion without supplemental feed, in addition to representing soil ingestion on degraded pasture with supplemental feed. The New Zealand data also indicate that the soil ingestion range in this memo might underestimate soil ingestion without supplemental feed, but the applicability of range conditions in New Zealand to pastures along MFLBC is unknown. The use of soil ingestion on sparsely vegetated lots with supplemental feed as a surrogate for soil ingestion on pasture without supplemental feed therefore may have a fairly high degree of uncertainty, and possibly underestimates potential exposure. The mean soil ingestion range used in this memo is supported by reasonable consistency with the 2 % (dairy cattle) and 4 % (beef cattle) values used to calculate recommended mean soil ingestion quantities in the peer-reviewed Human Health Risk

Assessment Protocol for Hazardous Waste Combustion Facilities (§ 5.4.4.4 and 5.4.5.4 in USEPA 2005b).

Concern over un-supplemented grazing might be misplaced because supplemental feed is required for high-producing dairy cows to attain maximal milk productivity. However, even for dairy cows, supplemental feed will not necessarily reduce mirex uptake. If supplemental forage is harvested from contaminated fields, the supplement will have absorbed mirex. More significantly, pasture plants have relatively high levels of adhered soil (Fries, et al. 1982; Beresford and Howard 1991), so provision of supplemental feed harvested from contaminated pasture would likely not affect soil ingestion. As demonstrated by the clean supplemental feed scenario in this memo, provision of clean supplemental feed by itself has only a minor impact on soil PRGs because plant uptake of mirex is a small contributor to cattle exposure in comparison to soil ingestion.

Even though the best milk production is achieved with supplemental feed, its provision is an economic decision. To illustrate, as a result of European Union policies that resulted in surpluses of dairy products and meat, there has been less emphasis on intensive livestock rearing, and "less intensive production with reduced supplementary feed has meant that grazing livestock are now more dependent on grass as their main source of nutrition and thus become dependent on the mineral status of the farm and its soils" (Thorton 2002). So provision of supplemental feed is not guaranteed.

A confounding factor in the Fries, et al. (1982) study of cattle soil ingestion is that non-lactating cattle confined to buildings and paved lots with no direct access to soil exhibited twice the mean apparent soil ingestion as lactating cows similarly confined (0.7 % vs. 0.3 %, respectively), which was attributed to possible ingestion of wood shaving bedding in the single farm that confined non-lactating cattle. Similarly, in a separate set of farms, mean soil ingestion by non-lactating cattle with access to unvegetated unpaved lots was higher than that of lactating cows under the same conditions (1.3 % vs. 0.8 %, respectively). Both groups were provided "abundant" supplemental feed, but the authors stated that "the wide differences in [the supplied feed] diet that could affect animal's appetite for soil" may be responsible. In any case, the disparities within each group between apparent soil ingestion without access to soil and increased soil ingestion with access only to bare soil "indicates that some soil ingestion by cattle is an active process and not just a passive process associated with grazing" (Fries, et al. 1982). The uncertainties associated with the Fries, et al. (1982) study are unlikely to have a major impact on the outcomes of this memo because the potential magnitude is much less than that potentially associated with the uncertainty of underestimating un-supplemented soil ingestion.

The soil ingestion range selected in this memo is a range of mean values calculated as the average of multiple management unit mean values reported for each treatment (4 means for pasture, and 5 means for sparsely vegetated lots). For a more conservative approach, Fries, et al. (1982) reported the 95 % upper confidence limit for soil ingestion of cows was as high as 6 % in individual management units. This approach is not taken in this memo to avoid multiplication of conservative assumptions since the beef and milk targets are based on the reasonable maximum exposure (RME) scenarios (EA 2004).

Conclusions

Mirex soil PRGs range from 0.3-0.6 mg/kg for cattle grazing on degraded pastures with supplemental feed, which is also treated as a surrogate for cattle grazing without supplement feed, to 1.4-2.8 mg/kg for cattle grazing on productive pasture with supplemental feed (Table 1), based on risks associated with consumption of milk or beef products. Additional soil PRGs based on alternative risk estimates are shown in Appendices B.1 – B.3. For any single beef or milk target, there is more than a 3- to 4-fold range in soil PRGs that reflects uncertainty in two processes affecting cattle exposure to mirex – soil dietary fraction and plant bioaccumulation. There is large uncertainty over bioaccumulation of mirex in plants, but it results in much less uncertainty in soil PRGs because soil ingestion is the predominant exposure route of cattle to mirex. Cattle soil ingestion is well-constrained by a study of dairy cattle provided supplemental feed and grazed on a range of pasture conditions in Michigan. One source of uncertainty is that soil ingestion values for cattle without supplement feed in the Central or Eastern U.S. were not located. The soil ingestion range is supported by broad consistency with mean cattle soil ingestion in a recent U.S. EPA peer-reviewed protocol (USEPA 2005b). Uncertainty over biotransfer of mirex from diet to beef or milk is not reflected in the soil PRG ranges, because biotransfer is modeled in this memo with the best available approach. The regression model used in this memo for calculating biotransfer is indirectly supported by an independent deterministic model of contaminant uptake in cattle, both of which show similar predictive performance for stable chemicals. Empirical studies of mirex biotransfer in cattle are not available with up-to-date mirex analytical methods, so there is unavoidable uncertainty over the modeled mirex biotransfer values. Earlier alternative regression methods indicate higher mirex biotransfer than used in this memo, but the performance of the earlier methods has been shown to be much poorer in comparison with the selected biotransfer regression method. The soil PRGs presented in this memo do not represent worse-case scenarios for soil ingestion by cattle or mirex biotransfer to milk or beef, but are based instead on a range of plausible mean soil ingestion values and the best available mirex biotransfer estimate.

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Appendix A.1. Derivation of the Equation for Calculating Preliminary Remedial Goals for Pasture Soil from Target Mirex Concentrations in Beef or Milk.

[1] $C_{fat} = IR_{mirex} * BTF_{fat}$

C_{fat} – mirex concentration in beef or milk fat (mg mirex / kg fat)
IR_{mirex} – mirex daily intake rate to cows (mg mirex / day)
BTF_{fat} – biotransfer factor to fat = mirex concentration in beef or milk fat / mirex daily intake rate
= (mg mirex / kg fat) / (mg mirex / day) = day / kg fat

[2] $IR_{mirex} = FIR * (C_{forage} + (C_{soil} * DF_{soil}))$

FIR – daily food ingestion rate (kg forage dw / day)
forage – plants grazed by cows
dw – dry weight
C_{forage} – mirex concentration in forage (mg mirex / kg forage dw)
C_{soil} – mirex concentration in soil (mg mirex / kg soil dw)
DF_{soil} – dietary fraction of soil = (kg soil dw / day) / (kg forage dw / day) = unitless ratio (dry weight basis)

[3] $C_{forage} = C_{soil} * BAF$

BAF – mirex bioaccumulation factor in plants (aboveground portion)
= (mg mirex / kg plant dw) / (mg mirex / kg soil dw) = unitless ratio (dry weight basis)

Combine Equations 2 and 3:

[4] $IR_{mirex} = FIR * C_{soil} * (BAF + DF_{soil})$

Combine Equations 1 and 4:

[5} $C_{fat} = FIR * C_{soil} * (BAF + DF_{soil}) * BTF_{fat}$

Rearrange Equation 5:

[6] $C_{soil} = C_{fat} / (FIR * BTF_{fat} * (BAF + DF_{soil}))$

Change C_{fat} to beef or milk target ($C_{fat\ target}$), and C_{soil} to soil preliminary remedial goal (PRG):

[7] Soil PRG = $C_{fat\ target} / (FIR * BTF_{fat} * (BAF + DF_{soil}))$

Assumptions:
Cows graze only in potentially contaminated fields.
Cow drinking water is an insignificant source of mirex relative to dietary exposure.
Inhalation is an insignificant source of mirex relative to dietary exposure.
Supplemental forage is harvested from potentially contaminated fields (on-site source), or not provided.

Appendix A.2. Modification of the Equation for Calculating Preliminary Remedial Goals for Pasture Soil from Target Mirex Concentrations in Milk to Include Provision of Supplemental Clean Feed to Dairy Cows.

Modified equations from Appendix A.1:

[1] no change

[2] $IR_{mirex} = FIR * (C_{f+f} + (C_{soil} * DF_{soil}))$

C_{f+f} – mirex concentration in combined pasture forage and supplemental feed (mg mirex / kg forage and feed dw)

[3] $C_{f+f} = C_{soil} * BAF_{adj}$

[3b] $BAF_{adj} = BAF * DF_{pasture}$

BAF_{adj} – mirex bioaccumulation factor in combined pasture forage and supplemental feed
$DF_{pasture}$ – dietary fraction from grazing in contaminated pasture (kg pasture forage dw / kg forage and feed dw) = unitless ratio (dry weight basis)

[4] – [6] BAF_{adj} replaces BAF

[7] Soil PRG = $C_{fat\ target} / (FIR * BTF_{fat} * (BAF_{adj} + DF_{soil}))$

Assumptions:
Cows graze only in potentially contaminated fields.
Cow drinking water is an insignificant source of mirex relative to dietary exposure.
Inhalation is an insignificant source of mirex relative to dietary exposure.
Supplemental clean feed comes from an off-site source and has no mirex residues.

Appendix B.1. Beef-based Soil Mirex Preliminary Remedial Goals

BTF_{fat} (d/kg dw)		0.13	0.13	0.13	0.13	0.48	0.48	0.48	0.48
Plant BAF (dw ratio)		0.004	0.004	0.045	0.045	0.004	0.004	0.045	0.045
DF_{soil} (dw ratio)		0.0138	0.0377	0.0138	0.0377	0.0138	0.0377	0.0138	0.0377
	Beef Target	Soil Mirex PRGs (mg/kg)							
Basis	(mg/kg fat)								
10^{-6}	0.00516	0.3	0.1	0.08	0.06	0.07	0.03	0.02	0.02
10^{-5}	0.0516	2.8	1.2	0.8	0.6	0.75	0.3	0.2	0.16
HQ = 1	0.234	13	5.4	3.9	2.7	3.4	1.4	1.0	0.7
10^{-4}	0.516	28	12	8.5	6.0	7.5	3.2	2.3	1.6

Appendix B.1 Inputs and Sources

BTF_{fat} (biotransfer factor):

0.13 d/kg, calculated as $\log BTF_{fat} = -0.099 \log K_{ow}^2 + 1.07 \log K_{ow} - 3.56$ (RTI 2005)

Mirex $\log K_{ow}$ (octanol-water partitioning coefficient) = 6.89 (RTI 2005)

0.48 d/kg, calculated as $\log BTF = 0.525\ ^1X - 5.904$ (Dowdy, et al. 1996), and $BTF_{fat} = 10^{BTF}$ / beef fat content

Mirex ^1X (normal path first-order molecular conductivity index) = 9.5 (Dowdy, et al. 1996)

Beef fat content = 0.25 (Dowdy, et al. 1996, and Travis and Arms 1988)

Plant BAF (bioaccumulation factor):

0.004, calculated as $\log BAF = -0.578 \log K_{ow} + 1.588$ (Travis and Arms 1988)

0.045, calculated as $\log BAF = -0.204\ ^1X + 0.589$ (Dowdy and McKone 1997)

DF_{soil} (soil dietary fraction):

0.0138 to 0.0377, range for yearling and dry cows with supplemental feed on pasture or sparse vegetation lot (Fries, et al. 1982)

FIR (food intake rate):

8 kg/d, value for non-lactating cattle (Travis and Arms 1988; RTI 2005)

Target Basis:

10^{-4}, 10^{-5}, and 10^{-6} are estimated cancer risks.

HQ (hazard quotient) = 1 is an estimated non-cancer risk.

Beef Target = Exposure Medium RME Value / Cancer Risk or Non-Cancer Hazard Quotient (values provided by Sheila Abraham, OEPA, calculated from Table K-95 – Reasonable Maximum Exposure, Appendix K, Endangerment Assessment for the Nease Chemical Company 2004). 10^{-5} and 10^{-4} based targets are extrapolated from the 10^{-6} target. Note that the Exposure Medium RME Value in Table K-95 is for beef fat (mg mirex / kg fat).

Assumptions:

See Appendix A.1.

Appendix B.2. Milk-based Soil Mirex Preliminary Remedial Goals

BTF_{fat} (d/kg dw)		0.13	0.13	0.13	0.13	0.33	0.33	0.33	0.33
Plant BAF (dw ratio)		0.004	0.004	0.045	0.045	0.004	0.004	0.045	0.045
DF_{soil} (dw ratio)		0.0138	0.0377	0.0138	0.0377	0.0138	0.0377	0.0138	0.0377
	Milk Target	Soil Mirex PRGs (mg/kg)							
Basis	(mg/kg fat)								
10^{-6}	0.00505	0.14	0.06	0.04	0.03	0.05	0.02	0.02	0.01
10^{-5}	0.0505	1.4	0.6	0.4	0.3	0.54	0.23	0.16	0.12
HQ = 1	0.167	4.5	1.9	1.4	1.0	1.8	0.76	0.54	0.38
10^{-4}	0.505	14	5.8	4.2	3.0	5.4	2.3	1.6	1.2

Appendix B.2 Inputs and Sources

BTF_{fat} (biotransfer factor):

0.13 d/kg (RTI 2005), see Appendix B.1 Inputs and Sources

0.33 d/kg, calculated as log BTF = 0.421 ^{1}X - 5.879 (Dowdy, et al. 1996), and $BTF_{fat} = 10^{BTF}$ / milk fat content

Mirex ^{1}X (normal path first-order molecular conductivity index) = 9.5 (Dowdy, et al. 1996)

Milk fat content = 0.04 (Dowdy, et al. 1996; RTI 2005)

Plant BAF (bioaccumulation factor):

See Appendix B.1 Inputs and Sources

DF_{soil} (soil dietary fraction):

See Appendix B.1 Inputs and Sources

FIR (food intake rate):

16 kg/d, value for lactating cows (Travis and Arms 1988; Dowdy, et al. 1996; RTI 2005)

Target Basis:

See Appendix B.1 Inputs and Sources

Milk Target = Exposure Medium RME Value / Cancer Risk or Non-Cancer Hazard Quotient (values provided by Sheila Abraham, OEPA, calculated from Table K-96 – Reasonable Maximum Exposure, Appendix K, Endangerment Assessment for the Nease Chemical Company 2004). 10^{-5} and 10^{-4} based targets are extrapolated from the 10^{-6} target. Note that the Exposure Medium RME Value in Table K-96 is for milk fat (mg mirex / kg fat).

Assumptions:

See Appendix A.1.

Appendix B.3. Milk-based Soil Mirex Preliminary Remedial Goals with Supplemental Clean Feed

BTF_{fat} (d/kg dw)		0.13	0.13	0.13	0.13	0.33	0.33	0.33	0.33
Plant BAF (dw ratio)		0.004	0.004	0.045	0.045	0.004	0.004	0.045	0.045
Adjusted BAF (dw ratio)		0.001	0.001	0.012	0.012	0.001	0.001	0.012	0.012
DF_{soil} (dw ratio)		0.0138	0.0377	0.0138	0.0377	0.0138	0.0377	0.0138	0.0377
	Milk Target				Soil Mirex PRGs (mg/kg)				
Basis	(mg/kg fat)								
10^{-6}	0.00505	0.16	0.06	0.10	0.05	0.06	0.02	0.04	0.02
10^{-5}	0.0505	1.6	0.63	1.0	0.49	0.64	0.25	0.38	0.19
HQ = 1	0.167	5.4	2.1	3.2	1.6	2.1	0.82	1.3	0.64
10^{-4}	0.505	16	6.3	9.6	4.9	6.4	2.5	3.8	1.9

Appendix B.3 Inputs and Sources

BTF_{fat} (biotransfer factor):
See Appendix B.2 Inputs and Sources

Plant BAF (bioaccumulation factor):
See Appendix B.1 Inputs and Sources

BAF_{adj} (adjusted BAF – bioaccumulation factor in combined pasture forage and supplemental clean feed):
Calculated as Plant BAF * Dietary fraction from pasture forage (dw basis)

$DF_{pasture}$ (dietary fraction from pasture forage):
0.257 kg pasture forage dw / kg forage and feed dw = unitless ratio (dry weight basis) (Soder and Muller 2007)

DF_{soil} (soil dietary fraction):
See Appendix B.1 Inputs and Sources

FIR (food intake rate):
See Appendix B.2 Inputs and Sources

Target Basis:
See Appendix B.1 Inputs and Sources

Milk Target:
See Appendix B.2 Inputs and Sources

Assumptions:
See Appendix A.2